Response to reviewers

[Reviewer comments in blue font. Our responses in regular, black font. **Changes to manuscript in bold, black font**. Certain minor modifications – only figure labels and axes – that need to be made to the manuscript and will be done before final submission are in red, regular font.**]**

Response to Anonymous Referee #1:

"There is a lot of discussion mixed in with the results, which by itself is not a problem, but quite often I find the discussion that does exist in the results section to be unclear and unfocused.… In any case, my point is that the authors should attempt to better separate results from discussion. Also, the discussion section could probably benefit from the use of sub-headings just as in the results section."

We thank the referee for the extensive comments. Before moving on to specific responses, a general comment about the organization of the paper. Basically, in the results section, we presented data from our measurements, and following this in each sub-section is a presentation of comparable *results* from other studies. The discussion section then *discusses* possible reasons for differences between our results and previous studies. **However, to avoid confusion, we have moved the comparison results as well to the Discussion section, which is now organized as two parts: one for the BC/CO ratios and another for the BC MAC and mixing state.**

"Here, the authors seem to be arguing that Dickerson overestimated their background contributions (and thus also the BC/CO ratio), but then later seem to argue that there are legitimate reasons why the CO background in Mexico City might be quite different than that near the Indian sub-continent. Perhaps I am reading this incorrectly, but the language is ambiguous enough that it is difficult to tell."

We do not say that Dickerson et al. overestimated the background CO or even the BC/CO ratio; we just compare the MIRAGE data against the INDOEX data, as well as with other measurements; no judgment is meant or consciously implied. The referee is correct in understanding from the discussion that we believe there are multiple reasons why the background CO over Mexico should be different from that over the Indian Ocean.

"I found the rigor of the uncertainty analysis to be inconsistent. For example, although the authors provide uncertainty bounds on their SP2 BC measurements (+/- 22%), they make comparisons to other measurements without consideration of uncertainties. For example, with the BC/CO measurements, they state that their measurements are "similar" to some other measurements (2.5 ng/microgram vs. 1 ng/microgram), however I believe this is outside of the uncertainty of the measurements. What do the authors mean by "similar"? This is again a specific example, but this sort of issue (i.e. a lack of precision in the language used) is encountered throughout the manuscript. Also, it is not clear when the uncertainties reported by the authors are derived from propagation of errors or are the actual precision of the measurements (i.e. observed atmospheric variability)."

We have tried to provide uncertainty and bias estimates where possible, explicitly. Hence, the BC/CO ratio and the BC mass absorption cross-section are provided with propagated uncertainties from each measurement. This was stated as such for the BC MAC on line 15 of page 9093, though not for the BC/CO ratio; this has been rectified in the revision thusly:

"the mean BC/CO_{net} ratio for both flights is around 3.1 (ng.m^{-3}-STP)/ppbv, or ~2.5 ng-BC/µg-CO, with a **propagated measurement uncertainty** of about ±25%."

If a value is specified as average and standard deviation (SD), such as in the event-wise BC MAC data, that represents atmospheric variability, as the SD associated with an average should mean.

For the specific instance where we compare our BC/CO ratios with that from other studies, note that we do not only reference the Baumgardner et al. (2007) data, but also the compilation by Kondo et al. (2006). Thus, the effective range of BC/CO measurements is 0.88 to 6 ng/m^3/ppbv; our value of 3.1 (ng/m^3-STP)/ppbv is in the middle of this range, and hence, we consider the comparison "similar", which, as per Merriam-Webster, means "alike in substance or essentials."

However, the discussion has been modified as follows, since the Baumgardner et al. results are from Mexico City and more comparable than values from Tokyo and other cities:

"**Thus, our airborne BC/CO_{net} ratios of 3.1 (ng/m^3-STP)/ppbv or 2.5 ng/µg, corrected to near-surface STP conditions, are similar to these surface measurements, though higher than the Baumgardner et al. SP2 measurements in Mexico City. One possible reason for this discrepancy is that the Baumgardner et al. measurements were close to vehicular traffic, while the C130 measurements include other source contributions, particularly mountain fires.**"

"Of perhaps more serious concern to me with respect to uncertainties or limits of the system is the discussion of particle ageing (Figures 8 and 9). Firstly, the authors show size distributions as a function of scattering/incandescence lag-time that go from ca. 65 nm to 400 nm. However, it is stated clearly (and Figure 2 indicates) that the data are not to be trusted below ca. 145 nm or above 330 nm. Thus, the data shown in these figures should be truncated accordingly or the authors must very carefully justify the inclusion of data outside of this range. However, much of the "interesting" data in Figures 8 and 9 is right around 145 nm and the conclusions are to some extent based on shifts within this region. The ageing of particles should be re-considered in the appropriate experimental framework (i.e. within the limits of the system)."

We have not explicitly stated that the SP2 data below 145 nm is not to be trusted. On page 9087, Lines 15-18, we state:

"Though the SP2 can detect a BC mass of 1 fg or lower, calibrations for the MIRAGE experiment were made only down to about 3 fg-BC, or about 145 nm MED. The mass limitation at the upper end and the mass and count limitations at the lower end mean that the SP2 does not measure the full range of atmospheric BC."

We should specify that the “count limitations at the lower end” does not imply that 145 nm is the lower limit of “good” data. The 145 nm value was a limitation of the material then used for calibrating the SP2, Alfa glassy carbon (we were not able to get good DMA peaks below this value.) Later work with Aquadag and fullerene soot make us confident in the linearity of the SP2 down to at least 1 fg (which is key), but since these measurements were made after MIRAGE and INTEX-B with some changes in the detector gains, we cannot use these calibrations with the MIRAGE data. We have modified the manuscript as follows:

“Though the SP2 can detect a BC mass of 1 fg or lower, calibrations for the MIRAGE experiment were made only down to about 3 fg-BC, or about 145 nm MED, due to limitations on the material, Alfa glassy carbon, used to calibrate the SP2 during MIRAGE. Later calibrations with better soot proxies including Acheson Aquadag™ (a synthetic graphite) and fullerene soot have shown the linearity of the SP2 response down to 1 fg-BC (100 nm MED), indicating the validity of the data down to this size.”

We clearly do not dispute that the SP2 does not detect all particles, as Figure 2 shows and the section on SP2 scaling efficiency explains at some length. However, the key indicator in Figures 8 and 9 (Figures 12, 13 in the revision) ***is the shift in the incandescent lag***. Whether a particle is thickly-coated or thinly-coated, the SP2 response to that particle is practically the same, as noted in Slowik et al. (2007).

“I also find the particle ageing discussion to be unnecessarily qualitative. The authors claim that there is “an apparent increase in the modal BC MED (BC mass/particle) of the thinly-coated BC mode” for aged vs. fresh particles. The authors can presumably determine log-normal fits during these periods from the data. They should do this and explicitly report the number based mean and median diameters along with uncertainty bounds. In other words, there is no reason that this increase needs to be “apparent” when it can be quantified.”

The referee’s comment is well-taken, and this was an oversight on our part. Subsequent examination of the data as recommended by the reviewer suggests that shifts in the modal MED of thinly-coated BC are not significant, and **we have removed this result and the corresponding discussion from the revision**.

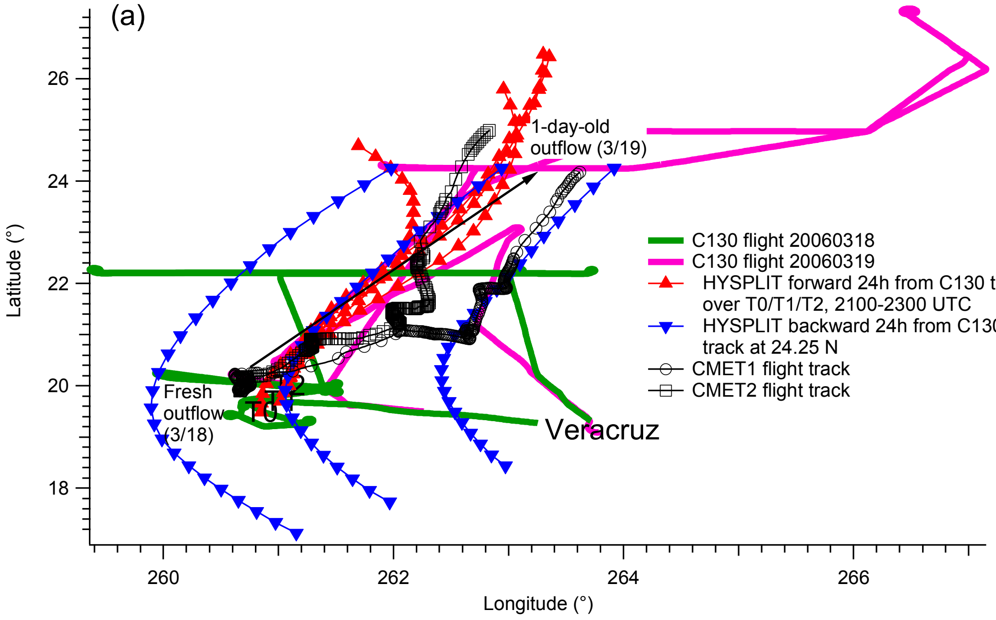
“Figure 8, bottom left and bottom middle panels: : :as an aside, it would help if these were labeled a, b, c, etc.).”

The revision will incorporate formal axis labels, as well as panels labels as (a), (b), (c) etc., which is easier than wordy positional descriptions.

“Of further concern with the particle ageing studies is that the largest change in the thickly vs. thinly coated fraction does not appear to derive from particle ageing, but from a change in altitude sampled on a given day… If such large changes in the thick vs. thin coatings can be observed just from changing altitude, how can the authors be confident that they are indeed encountering the same air mass one or two days later? It is argued that they knew where to sample from chemical forecasting, but have they attempted to track the sampled air masses back in time to provide confidence that they are sampling the same air masses? This observation is only very briefly noted at the very end of the manuscript but should be dealt with more prominently if the authors are going to provide confidence in their conclusions."

The difference between the BC aerosol characteristics at 3.5-3.8 km ASL and at 4.1 km ASL is not due to differences in altitude, **but because these are two different layers, which has been confirmed by WRF-FLEXPART modeling**. The lower layer at 3.5-3.8 km above sea level is fresher Mexico City outflow, while that at 4.1 km ASL is older outflow. The SP2 incandescent lag-size distributions do indeed suggest that the layer at 4.1 km is older with more thickly-coated BC, while the lower layers are dominated by thinly-coated BC.

We have used HYSPLIT to determine forward and backward trajectories (as we mentioned toward the end of the manuscript), but found the results less than satisfying, which is why we did not give it more consideration in the manuscript. For instance, HYSPLIT does not consider particle dispersion, which is an important factor in determining the air parcel position. The figure below shows one such example, where the HYSPLIT forward and backward trajectories are shown for March 18 and 19 respectively. Per HYSPLIT, the age of the plume at 24.25 °N is much shorter than one full day, and includes aerosol from a broader spatial range than just Mexico City (though Mexico City is likely the dominant contributor.)



However, the HYSPLIT trajectories are contradicted by the Controlled METeorological (CMET) balloons used on March 18 and 19 to track Mexico City outflow which originated on March 18. The NCAR C130 sampled 1-day-old MCMA outflow between the CMET balloons on March 19, and this is as direct evidence as possible that the plume at this location (24.25 °N) was indeed 1-day-old Mexico City pollution.

The revision includes an extensive discussion of plume age using HYSPLIT trajectories, WRF-FLEXPART modeling, and CMET balloon tracks.

"The authors present data in Figure 4 on BC concentration measurements for four days. However, in Figure 5 (BC/CO) and Figure 6 (absorption cross-section) they only consider two of these days (note: the dates are not given in Figure 6). Why are the flights on the other dates not considered in Figures 5 and 6? Presumably they would contribute further information. The same goes for Table 2.

In Table 1 the authors present average BC concentrations for different time periods during the individual flights. It would be useful if they were to also include columns for the average BC/CO and MAC during these periods, along with the appropriate uncertainties."

Table 2 and Figures 5 and 6 concentrate on data from March 18 and 22 because the March 19 and March 23 flights did not show a significant difference from the other two days. However, as per the referee's comment about including BC/CO and MAC in Table 1, **the revision incorporates Table 2 into Table 1, thus presenting the BC/CO and MAC data from the March 19 and 23 flights in tabular form.**

"The discussion concerning the BC/CO ratios as they compare to other measurements is insufficient. First, the authors state "Figure 5a shows a substantial CO concentration apparently not associated with BC." It is not visibly clear what is meant by this. Although they reasonably argue that some of the CO in the sampled air masses is old, it is not clear how this can be discerned simply from looking at Figure 5a. Second, the authors state that a comparison with Baumgardner et al. is probably acceptable to within a factor of two. It needs to be made clearer why this is the case. Third, the discussion with respect to the Dickerson results is confusing, as noted above. Related to this, the authors state that the BC/CO derived here is "comparable to corrected MOPPITT data over the Pacific Ocean off the Mexican coast for March/April 2006." I assume they mean their estimate of background CO, not BC/CO. Also, much of the discussion given relating to the BC/CO is focused not on BC/CO, but on proper estimation of the background CO. Based on their data the authors should be able to provide a reasonable estimate of how a poor estimate of background CO would influence their results. In any case, the discussion of specific CO emissions from different regions of the world has little bearing on the BC/CO; it is the relative emissions of BC and CO that will be of most importance."

The statement that there is a substantial CO concentration apparently not associated with BC just means the existence of background CO, which, presumably, is older than BC and CO emissions from, say, Mexico City (and hence, called "background CO.") The discussion of differences between BC and CO lifetimes is provided as explanation for the existence of background CO, and as way to explain Figure 5a, and more specifically, the CO-intercept in Figure 5a, not the other way around.

The MOPITT reference comes in a paragraph (lines 12-20, page 9092) that explicitly starts with the words “the background CO”, and there is no mention of the term “BC/CO” in the paragraph; the referee is mistaken.

We have not found a significant effect of issues with estimation of background CO on the BC/CO ratio. The discussion on specific sources of CO from Mexico and the Indian subcontinent was in reference to the factor-of-two difference between the CO background we measure for MIRAGE and the results of Dickerson et al. for INDOEX. The referee’s comment about differences in relative emissions of BC and CO is relevant to differences in the BC/CO ratio. However, given that BC and CO are both formed by incomplete combustion from different sources, what determines the BC/CO ratio is the source mixture, as we mention in the discussion section:

“One explanation for the lower BC/CO ratios over Mexico compared to the INDOEX data is that the source mix may be different between the Indian subcontinent and Mexico. Baumgardner et al. (2002) suggest that operating conditions including the altitude can also affect the BC/CO ratio.”

The statement about the SP2-based BC/CO being comparable to the thermal-optical EC-based ratios compiled by Kondo et al. to within a factor-of-two was based on uncertainties with EC measurements being generally within a factor-of-two (comparing TOT and TOR methods, e.g. the works by Chow, and previous work by the first author of this study.) **However, this statement has been deleted.**

“Finally, I’ll consider the presentation and discussion of the mass absorption efficiency measurements. First, to state that the MAE for the biomass burning period is “somewhat higher” than the other periods is not true within the uncertainties of the measurement.”

The statement has been deleted.

“Second, I find the discussion of the influence of coatings on potential absorption “enhancements” to be much too simplistic. The authors state multiple times that the effect of ageing is to increase absorption by 50% over fresh values. As they are undoubtedly aware, this 50% value is only true for very specific combinations of core sizes and coating thicknesses. It can vary dramatically around this value. From Figure 1 I estimate the core diameter to be □190 nm and given the “typical” lag times the coating thickness to be □20 nm. For these conditions the expected enhancement (in a coreshell spherical particle Mie approximation) is only 20%, not 50%. My point here is that the authors have the data available to make their discussion much more quantitative and should attempt to move beyond broad (and potentially misleading) generalizations that ageing can simply be captured as a 50% enhancement in absorption.”

There are only two instances where the enhancement in MAC due to coating is stated simply as 50% (line 20, page 9093 and line 9, page 9098.) These refer *specifically to the MAC of aged BC compared to that of fresh BC*; as Bond, Habib and Bergstrom state in their abstract:

“we suggest that absorption by aged aerosol is about 1.5 times greater than that of fresh aerosol.”

The other instances use a variation of “by as much as 50%” – different from the blanket “by 50%” as the reviewer states. Clearly, as the reviewer indicates, there is a substantial fraction of BC particles that are apparently thinly-coated, which will have a lower absorption amplification; hence, the average MAC we measure is *within the upper bound of MAC for aged soot* ("by as much as 50%"), calculated as (7.5+1.2)*1.5 = 13.05 m2/g. We must also caution the reviewer that, as we have explicitly stated at the end of the section on "optical particle diameter", the coating thicknesses shown in Figure 3 (based up on which the reviewer estimates a coating thickness of 20 nm) are likely biased low. Given the chicken-and-egg nature of the problem (needing to know the non-BC coating thickness to determine the overall refractive index to determine the particle size and thus, the coating thickness), as well as the uncertainties with the coating thickness due to volatilization of the non-BC coating of incandescent particles (as explained in the manuscript), we refrain from finessing this portion of the data further.

"Third, I do not find the speculation about there potentially being a balance between mixing state, a bias in the PSAP absorption measurements and the absorption enhancement to be particularly strong and not necessarily supportable from previous observations. For example, Lack et al. makes no claim as to whether the PSAP bias depends on mixing state (only total organic relative to BC) and Cappa et al. actually used an external mixture yet still saw a significant bias in the PSAP measurements. Thus, it is equally plausible that coagulation would act to increase the absorption enhancement while having no influence on the potential PSAP bias, rather than leading to a cancellation of effects. Related to this, I believe the organic aerosol measurements by De Carlo et al. were made on the same platform as the SP2 and PSAP measurements. One could potentially use the De Carlo measurements along with the SP2 measurements to determine a time-series of the OC/BC ratio during each flight (rather than the single point consideration given in the manuscript), which Lack found to be a good predictor of the magnitude of the PSAP bias. Given the high organic loadings during MILAGRO, the uncertainty in the absorption measurements is potentially much larger than the 20% stated (as mentioned, the measurements may be biased high by 40-70%). If a larger uncertainty is taken into consideration (or more specifically, a positive bias), then the agreement between the results presented here and the fresh emissions estimates from Bond and Bergstrom are not so bad (although there are certainly reasons to think that a real difference might exist, as discussed by the authors)."

The manuscript, as the reviewer quotes, does mention that there could be a positive bias of 40-70% in the PSAP absorption data due to the organic coating of the PSAP filter fibers. The revision now includes this explicit statement:

"The empirical studies by Lack et al. (2008) suggest that in such conditions, the PSAP could measure as much as 40-70% higher absorption. **A positive bias of 50% in the PSAP data would bring the BC mass absorption cross-section into better agreement with the MAC values for fresh soot found by Bond and Bergstrom in their review**."

The speculation about the enhancement amplification due to coating/ageing balancing out the PSAP absorption artifact is based on the fact that the MAC measured in fresh emissions is not significantly different from that in aged emissions. If the PSAP absorption artifact continued to exist in aged emissions in addition to the absorption enhancement brought about by condensation during aging, the BC MAC in aged air masses would be higher, which is not the case.

The uncertainty in the absorption measurements is explicitly stated as 20% "*excluding any bias due to liquid organic matter*" (line 26, page 9089), which is a reference to the PSAP bias found by Lack, Cappa and co-workers (which is discussed extensively later in the manuscript).

"Finally, I would argue that the comparison of the MAE measured here to that derived by Baumgardner et al. is not a fair comparison. The Baumgardner MAE values were determined in two ways: (1) based on PSAP measurements and (2) based on Mie theory and the measured particle size distribution. Baumgardner et al. found that the PSAP measurements gave a MAE approximately a factor of two greater than the Mie theory results, yet here the results are only compared to the Mie theory results and not to the more directly comparable PSAP results. (Admittedly, Baumgardner et al. do not explicitly give a MAE from their PSAP measurements. However, since they compare the measured PSAP absorption to absorption predicted based on their Mie Theory derived MAE and observed BC mass, and the PSAP absorption was higher by a factor of 2.3, the MAE from the PSAP would be correspondingly higher by a factor of 2.3. Furthermore, it is not surprising that Baumgardner et al. determined a MAE from theory that is much smaller than estimates from measurements. Bond and Bergstrom (2006) show very clearly that, for whatever reason, Mie theory is seemingly incapable of predicting MAE's as large as are observed; c.f. Bond and Bergstrom (2006) Figure 9). As such, I would strongly encourage the authors to remove any comparison to Baumgardner et al, unless they change the discussion to include the more directly comparable PSAP-based MAE."

The comparison with the Baumgardner et al. data was included because that of the study's relevance to Mexico City and their use of an SP2 to determine BC mass. ***The 2.3x difference between the PSAP and the SP2/Mie theory-derived values in Baumgardner et al. was really a difference in Babs – the ensemble absorption, not the specific absorption.*** Here is the relevant portion from Baumgardner et al.:

"The Babs from the PSAP and SP2 are well correlated but the PSAP values are on average 2.3 times larger than the SP2. This difference is primarily a result of the lower and upper sizing limits of the SP2, as discussed previously."

In other words, the SP2 BC mass presented in Baumgardner et al. did not account for the scaling factor (which corrects for BC mass outside the SP2 detection limits), and hence the large difference ***in Babs – not in the mass absorption cross-section***. We can not know the PSAP-based MAC from Baumgardner et al.'s data, unless we perform calculations to determine the SP2 mass scaling factors for the Baumgardner et al. data, and then derive PSAP-based MAC – which is beyond the scope of the current manuscript.

Specific comments:

Figures 4,5,6,7: Any points that are truly "zero" should be removed from the graphs. They are distracting. Also, it is mentioned that data during ascents/descents is not to be trusted in an absolute sense and so this data should be removed from all of these plots.

Figure 4 is a time-series of BC mass concentration; the data have to be reported as-is, including zeros. Eliminating them gives a false impression that data is not available.

As for the data during ascents/descents, the averages (which, frankly, are most useful) are presented for level portions of the flight in Table 1, while the averages for the two profiles are presented with a caveat, which should suffice to warn against use of these data.

Figures 8 and 9: The axes labels are not acceptable.

Not clear what the reviewer considers "unacceptable" – labels were not shown in Figure 8 (Figure 12 in the revision) as a concession to visual clarity (but the labels were described in the caption), but Figure 9 (Figure 13 in the revision) does include these labels. The revision will includes axes labels on Figure 8 (#12 in the revision).

Figure 6: The MAC was never defined as "kLAC" in the text yet it is used to label the axes.

The figure label in the revision will be modified to read MAC.

Page 9083, L21: It is not clear why the particles must be refractory. How is this a necessary part of the definition?

The particles have to be refractory because otherwise they would volatilize well before the high temperatures indicative of incandescence (~4000 K, Schwarz et al., 2006).

P. 9084, L. 9: The authors seem to be overstating the benefits of the SP2 here. Can the data retrieved from the SP2 be used confidently on a particle-by-particle basis? Or are the collected single-particle data typically averaged over some time?

The spectra from individual particles can be clearly seen and are separately recorded as a distinct event, e.g. Figure 3. While we did not explicitly stated the lower concentration limits possible with the SP2, we have detected (in other campaigns) concentrations as low as a few ng/m3 with 1-second resolution – much lower than is possible with filter-based measurements. The revision now includes this modified statement:

"Since the SP2 measures individual particles, it has a much lower detection limit than conventional filter-based measurements, allowing for highly time-resolved data collection **of concentrations as low as a few ng/m^3**."

P. 9087, L. 4: I find this difficult to understand. What do the authors specifically mean by "adds back 18% of the BC mass"?

The incandescent temperature ratio filter removes 22% of the BC mass; the shape-based criteria restores or "adds back" 18% of this removed BC mass, limiting the removal to just 4% of the mass. Not sure how that is hard to understand.

P. 9088, L. 3: I would recommend including a table that shows the log normal fit parameters and scaling factors as determined for each flight.

We can include such a table if required, though in our opinion it would not add much to the discussion ; the revised paper is quite lengthy as-is.

P. 9088, L. 15: More detail is needed here, such as the RI's assumed for the BC and non-BC material and a clearer discussion of the mixing rules employed.

The text has been modified as follows:

“The optical diameter calculations here assume a homogeneously-mixed BC:organosulfate mixture with BC volume fractions of 25%, 50%, 75% or 100%, whichever is closest to the estimated particle composition (see below for a limitation on this particular SP2 dataset that renders further refinement unnecessary). The refractive index of BC is assumed as 1.95-0.79i based on Bond and Bergstrom (2006), while that of the non-BC coating (organic matter/ammonium sulfate) is taken as 1.48-0i. A core-shell model does not make an appreciable difference in this calculation.”

Further detail is, we believe, not warranted given that the results from this calculation are only used to provide a qualitative indication of the relation between the incandescent lag and the coating thickness.

P. 9090, L. 5: What is the time-resolution on the reported data?

The figure and data are presented as 10-second averages, and the text has been modified as follows:

“The time histories of BC mass concentrations measured during these flights are presented in Fig. 4 **as 10-second averages** (normalized to STP, 1 bar and 273.15 K)”

P. 9090, L. 11: It is not clear why only the flight on March 18 is discussed here. I thought there were four flights being considered?

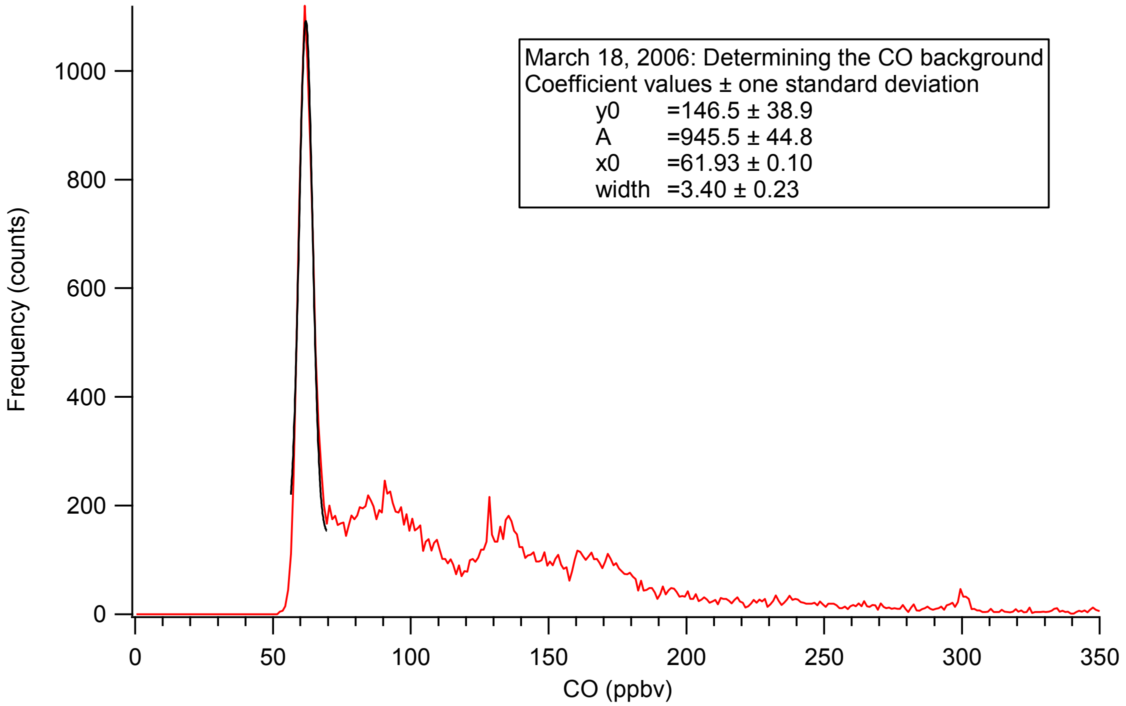
The description of the March 18 flight is just provided as an example; adding three more paragraphs describing in words what is already given in Figure 1 and Table 1 is, we believe, non-essential and would make for tedious reading.

P. 9091, L.18: It is not clear what is meant by the lowest frequency peak. I would recommend showing something more explicit.

The lowest frequency peak refers to the histogram of CO values, shown below, and as described in the manuscript thusly:

“For the March 18 and 22 flights, this value is estimated at 62 and 67 ppbv-CO respectively, determined rather simplistically by plotting a frequency distribution of the CO data, and picking the lowest frequency peak as the background CO concentration.”

While a picture may be worth a thousand words, the above description captures the essence of this procedure succinctly, and so we would prefer not to add another plot to the paper.



P. 9091, L.21: I might argue that this is a judgment call. One could reasonably look at the figures, as presented, and conclude that there is actually a lot of variability in the BC/CO ratio. Since the focus here seems to be on the average BC/CO values observed in different air masses being approximately the same, perhaps the data could be binned to show averages for specific regions (with uncertainties).

The revised Table 1 includes average BC/CO ratios (and standard deviations) for the different plumes.

P. 9099, L.15: It is not clear whether this is meant to be a general statement or specific to these measurements. If it is specific, the authors should consider whether or not it rained. If it did not rain during the measurement period, then wet removal processes would have had no influence on the observations.

The revision includes this statement about the March 23 data set:

"MODIS satellite images taken on March 23 at 20:00 UTC show rain clouds over the Gulf of Mexico, which suggests wet-scavenging of likely-hydrophilic thickly-coated BC at least on this day."

However, cloud images do not by themselves mean the aerosol passed through cloud or a rain event; thus, this is more a general statement.

Nit-picky comments:

Data are plural.

Not sure what this refers to. An example from the text, which shows data is considered plural:

“The MIRAGE CO data have a 3 ppbv precision.”

p. 9097, Line 3: This is a hypothesis, not a theory.

Fixed.

Response to Anonymous Referee #2:

“The discussion of MAC has little relevance to the other two issues.” – we strongly disagree. The mixing state of BC is very relevant to BC mass absorption cross-section (MAC) and consequently to aerosol radiative forcing, as numerous modeling studies by Bond, Fuller et al. have pointed out. In our manuscript, we have discussed the MAC results in light of the results on BC mixing state obtained using the SP2 (a data feature that is perhaps unique to the SP2). **To make this connection clear, the discussion is re-organized under two sub-headings: “Carbon monoxide and BC” and “BC mixing state and mass absorption cross-section.”**

Another point to note is that all three physical quantities discussed in the manuscript – BC/CO ratio, BC MAC and BC mixing state – are of relevance to global modeling work. The (additional) common thread connecting all three is the BC measurements from the SP2.

Comment 1: We do not conclude “no significant change of BC coating proceeded by aging.” In fact, the section on mixing state and Figures 8 and 9 (Figures 12, 13 in the revision) show these changes (the transition from thinly-coated BC to thickly-coated BC), and are explained as such. What we do point out is that BC MAC does not appear to change, in contrast to expectations based on modeling work by Bond, Fuller et al., and explore reasons why this is the case.

Whether the aging is by chemical transformation of organic aerosol present on the BC (when thinly-coated) or (more likely) condensation of semivolatile matter/sulfate formation is somewhat irrelevant to the discussion, as the SP2 cannot distinguish between inorganic and organic coatings. We do not claim to be able to do so, either.

[Figure 3 and the section on mixing state are presented solely to help the reader understand Figures 8,9 (12,13 in the revision).]

Comment 2: The conclusions from Kondo et al. as reported by the reviewer are in line with our own results. We have an extensive discussion of potential issues with the PSAP measurements, supported by experimental evidence and modeling results.

Though the referee considers filter-based measurements of aerosol absorption outdated, that is the method widely used even these days for aerosol light absorption (for example, in the NOAA ESRL/GMD network), not to mention the basis of historical datasets, and hence worthy of study. I would caution the reviewer against using these filter-based absorption measurements for BC mass measurements – if one finds absorption values unusable, then the BC mass derived from these same absorption measurements are of dubious value.

[Of course, as a scientist with a for-profit company that makes the only commercially-available *in-situ* aerosol absorption measuring instruments, the lead author would prefer everybody use a photoacoustic aerosol spectrometer, but this manuscript is not the place for a sales pitch.]

Comment 3: Trying to relate the BC/CO ratios to source mixtures is beyond the scope of this manuscript. However, this data is useful to global modeling work (as are the BC MAC and BC mixing state), and hence we compare our results to data obtained from Mexico City as well as other locations (including the Dickerson et al. work), to give some context to our results.